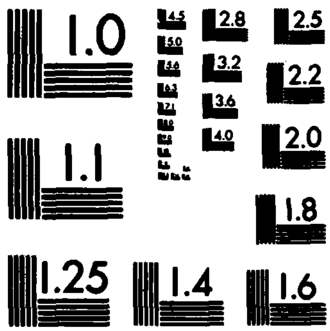
AD-A133 861 THE ANOMALOUS MAGNETORESISTANCE OF GRAPHITE AT HIGH MAGNETIC FIELDS(U) MASSACHUSETTS INST OF TECH CAMBRIDGE G TIMP ET AL. MAY 83 AFOSR-TR-83-0760 F49620-83-C-0011 1/1

UNCLASSIFIED F/G 20/3 NL

END

FILMED

DTIC



MICROCOPY RESOLUTION TEST CHART
NATIONAL BUREAU OF STANDARDS-1963-A

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered) Unclassified

AD-A133 861

REPORT DOCUMENTATION PAGE

READ INSTRUCTIONS BEFORE COMPLETING FORM

1. REPORT NUMBER AFOSR-TR- 83-0760	2. GOVT ACCESSION NO. AD-A133 861
3. RECIPIENT'S CATALOG NUMBER	
4. TITLE (and Subtitle) THE ANOMALOUS MAGNETORESISTANCE OF GRAPHITE AT HIGH MAGNETIC FIELDS	5. TYPE OF REPORT & PERIOD COVERED REPRINT
	6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) G. TIMP, P.D. DRESSELHAUS, T.C. CHIEU, G. DRESSELHAUS, Y. IYE	8. CONTRACT OR GRANT NUMBER(s) F49620-83-C-0011
9. PERFORMING ORGANIZATION NAME AND ADDRESS Massachusetts Institute of Technology 77 Massachusetts Avenue Cambridge, MA 02139	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 61102F 2306/C3
11. CONTROLLING OFFICE NAME AND ADDRESS Air Force Office of Scientific Research/NE Bolling AFB, DC 20332	12. REPORT DATE May 1983
	13. NUMBER OF PAGES 17
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)	15. SECURITY CLASS. (of this report) Unclassified
	15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Approved for public release; distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

Phys. Rev. (accepted) pages unavailable

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

anomalous magnetoresistance of graphite
high magnetic fields
charge density wave instability
angular dependence of magnetoresistance

20 83

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

See Over.

DTIC FILE COPY

DD FORM 1 JAN 73 1473 Unclassified

20.

The angular dependence of the high field magnetoresistance anomaly in graphite is reported. The observed angular dependence is consistent with the functional form for the phase boundary of a charge density wave transition suggested by Yoshioka and Fukuyama. Sharp periodic features in the resistivity occurring after the onset of the anomaly are reported.

The Anomalous Magnetoresistance of Graphite at High Magnetic Fields

G. Timp, P. D. Dresselhaus, T. C. Chieu
Department of Electrical Engineering and Computer Science and
Center for Materials Science and Engineering
Massachusetts Institute of Technology, Cambridge, MA 02139

G. Dresselhaus
Francis Bitter National Magnet Laboratory
Massachusetts Institute of Technology, Cambridge, MA 02139

and Y. Iye
Bell Laboratories, Murray Hill, NJ 07974

Abstract

The angular dependence of the high field magnetoresistance anomaly in graphite is reported. The observed angular dependence is consistent with the functional form for the phase boundary of a charge density wave transition suggested by Yoshioka and Fukuyama. Sharp periodic features in the resistivity occurring after the onset of the anomaly are reported.

PACS index: 72.15.Gd, 72.15.Nj

Introduction

The high field magnetoresistance anomaly in pristine graphite[1,2] has been examined in detail in order to further test the applicability of the theoretical model for this high magnetic field phase transition proposed by Yoshioka and Fukuyama.[3] According to their model, the high field magnetoresistance anomaly is associated with the formation of an electronic charge density wave (CDW) with an incommensurate c-axis periodicity. The predictions of this model for the electronic phase transition are consistent with the observed temperature dependence of the critical field for the magnetoresistance anomaly.[1,2] This theory was recently extended by Yoshioka[4] to calculate the dependence of the electronic phase transition on the angle θ between the magnetic field direction and the c-axis. The present work is directed toward providing an experimental test for the predicted angular dependence. Earlier experimental work[2] indicated that unresolved fine structure was also associated with the occurrence of the magnetoresistance anomaly. In the present work, the unusual properties of this fine structure (which is periodic in magnetic field H) is examined in more detail.

To investigate these unusual high field phenomena, we have used very carefully selected crystals of kish graphite. Our results on the magnitude of the magnetoresistance anomaly and the additional fine structure associated with this anomaly suggest that an additional mechanism may play a role in this high field regime.

Experimental

The kish graphite samples were prepared by precipitation from molten Ni and were later purified by heating in either F_2 or Cl_2 gas.[5] Though kish graphite is classified as a single crystal form of graphite, actual kish graphite samples used in these experiments normally contain several crystallites within their sample volume. Each crystallite typically shows a c-axis coherence distance of ~ 1000 Å and resolution-limited (hk0) x-ray line widths, indicating basal plane coherence distances in excess of 1 μm. A highly effective characterization method for selecting samples that show a large high field magnetoresistance anomaly was the identification of samples exhibiting large Shubnikov-de Haas oscillations in the low field range (below 8 T).

The graphite crystals were polygonal but irregular in shape. To avoid sample damage, the crystals were not trimmed. The transverse magnetoresistance was measured using a van der Pauw geometry;[6] the length to width ratio of typical samples was ~ 3. The contacts were made using a conducting silver epoxy. Under typical conditions, the DC resistance measurement was taken at a constant current of 100 μA. The voltage was detected by a Keithley 140 DC amplifier; the data acquistion was by computer. The magnetic field resolution required to observe the fine structure associated with the magnetoresistance anomaly is $(\Delta H/H) \sim 0.1$ T/25 T or about 0.4 %. Thus, for typical magnetic field sweep rates (10 T in 10 minutes) the detection circuit response time should not exceed 100 msec.

The transverse magnetoresistance was measured as a function of angle θ between the magnetic field and the c-axis of graphite. The design of the sample holder was such that θ could be varied while the samples remained essentially centered in the magnet. Expected magnetic field variations due to variations in sample position are less than 0.1% of the total field. The angle θ was determined from the known angular dependence of the SdH oscillations in graphite;[7] specifically, the 7.25 T spin-split cross section (associated with the Landau level $n = 0$, $\sigma = +$) was used for calibration. The sample holder position corresponding to $\theta = 0°$ was found by minimizing the value of magnetic field associated with the $n = 0$, $\sigma = +$ Landau level crossing of the Fermi surface.

A transverse magnetoresistance geometry was maintained so that the current in the samples was always perpendicular to the magnetic field, independent of angle. Magnetic fields up to 29 T were made available by using a hybrid magnet.[8] The hybrid magnet consists of an outer superconducting magnet generating a field from 5 T up to 7 T and a water-cooled Bitter magnet supplying a field which is variable from 0 to 22 T. The diameter of the magnet bore is 31 mm.

A ^{3}He evaporation refrigerator was used to attain low temperatures from 1.9 K down to 0.5 K. The samples were directly immersed in ^{3}He liquid. The temperature was determined by measuring the saturated vapor pressure of ^{3}He with a diaphragm-type pressure gauge (an MKS Baratron).

Results

The galvanomagnetic properties of graphite in the quantum limit have been previously examined.[9-22] The results can be summarized with reference to Fig. 1 which shows the transverse magnetoresistance at $\theta = 0 \pm 2°$ as a function of magnetic field. Below 7.3 T, Shubnikov-de Haas (SdH) oscillations are observed corresponding to two distinct extremal Fermi surface cross sections at 4.8 T (hole pocket) and 6.6 T (electron pocket).[9-10] The saturation and the negative slope of the magnetoresistance above 12 T have been associated by Iye et al.[2] with a linear increase in carrier concentration with increasing H.[11] The anomalous increase observed in the magnetoresistance for fields greater than 22 T has been studied experimentally by Tanuma et al.[1] and Iye et al.[2] The onset of the anomaly, defined by H_c (indicated by the arrow in Fig. 1), shows a striking temperature dependence which is apparently inconsistent with a single particle description of the magnetoresistance. Because of the high static field capability and crystal quality, we were able to resolve additional oscillatory features in the magnetoresistance at fields greater than the onset of the anomaly, as discussed in detail below.

The temperature dependence of H_c, the onset of the anomaly, observed in this work is summarized in the inset of Fig. 2. H_c was estimated from the maximum in the slope of resistivity as a function of magnetic field. The results for the "phase boundary" of H_c vs T_c measured here (represented by points) agree within experimental error with previous measurements (solid curve)[1,2] represented by the functional form:

$$T_c = \tau_c \exp[-\eta_c/H_c] \qquad (1)$$

in which $\tau_c = 94.9$ K and $\eta_c = 112$ T. As proposed by Yoshioka and Fukuyama,[2] the development of a gap at the Fermi level due to the Coulomb interaction between carriers would be manifested by a thermally activated transverse magnetoresistance with a phase boundary given by the dashed curve in the inset to Fig. 2. On the basis of this comparison, an identification of the high field magnetoresistance anomaly in graphite was made with the charge density wave instability model of Yoshioka and Fukuyama.[1,2]

To provide a further test for this identification, Yoshioka extended the model to consider the angular dependence of the charge density wave (CDW) phase transition. For the angular range accessible in the present experiment, his calculation of the angular dependence of H_c predicts that at constant temperature, $H_c(\theta)$ is approximately given by

$$H_c(0) \; / \; H_c(\theta) = \cos(\theta) \tag{2}$$

This result is physically reasonable since only the projection of the field onto the c-axis of graphite is expected to contribute to the CDW instability.

Figure 3 shows high field magnetoresistance measurements at several values of θ for the same sample as in Fig. 1, at the same temperature $T = 0.66$ K, and at the same value of the sample current $I = 100\ \mu A$. The data show that at constant temperature and current, the magnetic field onset of the magnetoresistance anomaly H_c increases with increasing θ. At the highest available field of 28.5 T, the magnetoresistance anomaly could not be seen beyond $\theta = 37.9°$.

Measurements of the angular dependence of H_c were made on 4 samples for a variety of temperatures and current levels. The results indicate that H_c is not dependent on the current level, at least for $I \leq 250\ \mu A$ (higher current levels caused some ohmic heating). The functional dependence of H_c on θ at constant T was found to be similar for all 4 samples that were investigated. Results for $H_c(0)$ / $H_c(\theta)$ are plotted in Fig. 2 as points for all samples, temperatures and current levels. The error associated with the estimate of H_c increases with increasing angle from 0.25% at $\theta = 0°$ to 1% at $\theta = 40°$ because the onset of the transition is less abrupt as the angle increases. The error arising from the determination of θ is less than 2°.

The experimental points fit well to a cos θ dependence (solid curve), in agreement with Yoshioka's model for this range of θ.[4] These results provide additional support to the identification of the high field magnetoresistance anomaly with the charge density wave electronic transition. We note that the angular dependence of H_c is similar to the angular dependence of the 7.25 T SdH oscillations. We further note that the region of saturation in the magnetoresistance below H_c and the region of negative slope beyond H_c show an angular dependence that is distinct from that of the anomaly.

The most striking and novel features discovered in these experiments are the periodic variations in the resistance which occur (Fig. 4) only above the onset of the mangetoresistance anomaly ($H > H_c$). Figure 4 shows an expanded view of the magnetoresistance record in Fig. 1 for $H > 20$ T, $I = 500\ \mu A$, $\theta = 0°$. The amplitude of the oscillations shows a phase change of π near the magnetic field, H_m, corresponding to the maximum of the anomaly. The oscillations are periodic in H and have been reproducibly observed in two of the five samples investigated.

The non-oscillatory component of the signal was estimated from a lowpass digitally filtered version of the record shown in Fig. 4a where the magnetoresistance anomaly exhibited no structure. The oscillatory features were obtained by subtraction of this non-oscillatory estimate from a synchronized version of the original resistance data record. The data records were synchronized using a linear interpolation scheme over the magnetic field range of interest.

The resulting oscillatory structure associated with the anomaly, shown as Fig. 4b, confirms the periodicity in magnetic field and the lineshape reversal of the fine structure near H_m noted above. Because the variation in the resistance appeared periodic in H, the data were analyzed using a standard discrete Fourier transform algorithm. (The data were also transformed assuming a periodicity in 1/H, and no appreciable frequency components were observed.) The data record, for the oscillatory component was digitally filtered to reduce side lobe contributions in the corresponding transform arising from the rectangular window function. The resulting power spectrum yields a fundamental period of 0.16 T, with second and third order harmonic content; the corresponding periods are 0.079 T and 0.056 T in second and third order, respectively. A periodicity of 0.17 T was observed in another sample which exhibited the fine structure in the resistance anomaly. Because of systematic errors associated with the magnet calibration and the number of periods counted, the difference between the two fundamental periods observed in different samples is not significant.

The data in Fig. 1 suggest that the magnetoresistance can be considered as the sum of a non-oscillatory, $R_{NOS}(H)$, and an oscillatory, $R_{OS}(H)$, contribution

$$R(H) = R_{NOS}(H) + R_{OS}(H) \tag{4}$$

in which $R_{NOS}(H)$ is associated with the one electron contribution from electrons in band states scattered from phonons or defects, and with the mechanism responsible for the magnetoresistance anomaly and $R_{OS}(H)$ is the corresponding contribution which is periodic in H.

Analysis of the amplitude of the oscillatory component (Fig. 4b) indicates that at constant current, $R_{OS}(H)$ has the functional form

$$R_{OS}(H) = r_{OS}(H)\ \{H/R\ \partial R/\partial H - 1\}\ \theta(H - H_c) \tag{5}$$

in which the step function $\theta(x) = 1$ for $x > 0$ and $\theta(x) = 0$ for $x < 0$. The oscillatory factor $r_{OS}(H)$ can be approximated as a sum of δ-functions,

$$r_{OS}(H) = \sum_{\ell} \delta(g\mu_B H - \ell\varepsilon_o) \tag{6}$$

where the g-factor g is taken as 2, μ_B is the Bohr magneton, and the energy $\varepsilon_o = 2.0 \times 10^{-5}$ eV is found from a fit to the experimental results of Fig. 4b, and is given by the dotted curve. It is of interest that ε_o is of the order of the spin-orbit splitting of the graphite π-bands. Finally, the envelope function $\{H/R\ \partial R/\partial H - 1\}$ depends on $R(H)/H$ and its derivative. We note that $R(H)/H$ is related to the variable E/H, the ratio of the electric to magnetic fields in the sample, since for the constant current conditions of this

experiment $E/H = (I/L)\ [R(H)/H]$ where I is the constant current and L is the length of the sample. The empirical dependence of the amplitude of the oscillations in Fig. 4b on $R(H)/H$ and its derivative thus suggests that the oscillatory behavior depends on the variable (E/H).

We note that the wave functions for the Landau levels $\phi_n(y - y_o)$ in crossed electric and magnetic fields depend on E/H, through the center location y_o of the harmonic oscillator, $y_o = (hk_H/m^* - E/H) / \omega_c$. The variable E/H causes a displacement of y_o, as also occurs in the Laughlin theory of the Quantum Hall Effect.[22] These observations suggest that the mechanism for the oscillations periodic in H might be related to the in-plane quantization of the harmonic oscillator wave functions. This is in contrast to the suggested mechanism for the high field magnetoresistance anomaly, of a charge density wave instability along the c-direction.

ACKNOWLEDGMENTS

The authors wish to thank Dr. L. Rubin and Dr. B. Brandt and the staff at the Francis Bitter National Magnet Laboratory for technical assistance and advice. We thank Prof. P.A. Lee, Prof. J.W. McClure, Dr. P. Tedrow, Dr. D. Yoshioka, and Prof. M.S. Dresselhaus for valuable suggestions. The kish graphite material was kindly provided by Prof. H. Suematsu of the University of Tsukuba. We acknowledge AFOSR contract #F49620-83-C-0011 for support of this work.

References

1. S. Tanuma, Y. Onuki, R. Inada, A. Furukawa, O. Takahashi, and Y. Iye, in Physics in High Magnetic Fields, vol. 24 of Springer Series in Solid State Sciences, edited by S. Chikazumi and N. Miura (Springer, Berlin, 1981) p. 316.

2. Y. Iye, P.M. Tedrow, G. Timp, M. Shayegan, M.S. Dresselhaus, G. Dresselhaus, A. Furukawa, and S. Tanuma, Phys. Rev. B25, 5478 (1982).

3. D. Yoshioka and H. Fukuyama, J. Phys. Soc. Jpn. 50, 725 (1981).

4. D. Yoshioka, (private communication). The Yoshioka calculation assumes the functional form of Ref. 3 where $[H(\theta) \cos\theta]$ is introduced for the magnetic field and an anisotropic g tensor is used for the spin term in the Hamiltonian.

5. H. Suematsu, (private communication). S. B. Austerman, in Chemistry and Physics of Carbon, edited by P. L. Walker and P. A. Thrower (Dekker, New York, 1968), Vol. 4, p. 137.

6. L. J. van der Pauw, Philips Res. Repts. 16, 187 (1961).

7. D. E. Soule, J. W. McClure, and L. B. Smith, Phys. Rev. A134, 453 (1964).

8. M. J. Leupold, J. R. Hale, Y. Iwasa, L. G. Rubin, and R. J. Weggel, IEEE Trans. Magn. 17, 1779 (1981).

9. J. W. McClure and W. J. Spry, Phys. Rev. 165, 809 (1968).

10. P. R. Schroeder, M. S. Dresselhaus, and A. Javan, Proc. of the Int. Conf. on Semimetals and Narrow Gap Semiconductors, edited by D. L. Carter and R. T. Bate, (Pergamon Press, New York, 1971) p. 139; E. Mendez, A. Misu, and M. S. Dresselhaus, Phys. Rev. B21, 827 (1980).

11. J. Heremans, J-P. Michenaud, M. Shayegan, and G. Dresselhaus, J. Phys. C14, 3541 (1981); Y. Iye, J. Heremans, K. Nakamura, G. Kido, N. Miura, J-P. Michenaud, and S. Tanuma, J. Phys. Soc. Jpn. 52, 1692 (1983).

12. J. W. McClure, Phys. Rev. 119, 606 (1960).

13. M. Inoue, J. Phys. Soc. Jpn. 17, 808 (1962).

14. K. Nakao, J. Phys. Soc. Jpn. 40, 761 (1976).

15. G. Dresselhaus, and M. S. Dresselhaus, Phys. Rev. B21, 827 (1980).

16. D. Shoenberg, Philos. Trans. R. Soc. (London) 245, 1 (1952).

17. J. W. McClure, Phys. Rev. 108, 612 (1957).

18. N. B. Brandt, B. A. Kapustin, V. G. Karavaev, A. S. Kotosonov, and E. A. Svistova, Zh. Eksp. Teor. Fiz. 67, 1136 (1974) [Sov. Phys.-JETP 40, 564 (1975)].

19. J. A. Woollam, D. J. Sellmyer, R. O. Dillon and I. L. Spain, in Low Temperature Physics LT. 13 (Plenum, New York, 1975), Vol. 4, p. 358.

20. J. A. Woollam, Phys. Lett. A32, 115 (1970).

21. K. Sugihara and J. A. Woollam, J. Phys. Soc. Jpn. 45, 1981 (1978).

22. R. Laughlin, Phys. Rev. B23, 5632 (1981).

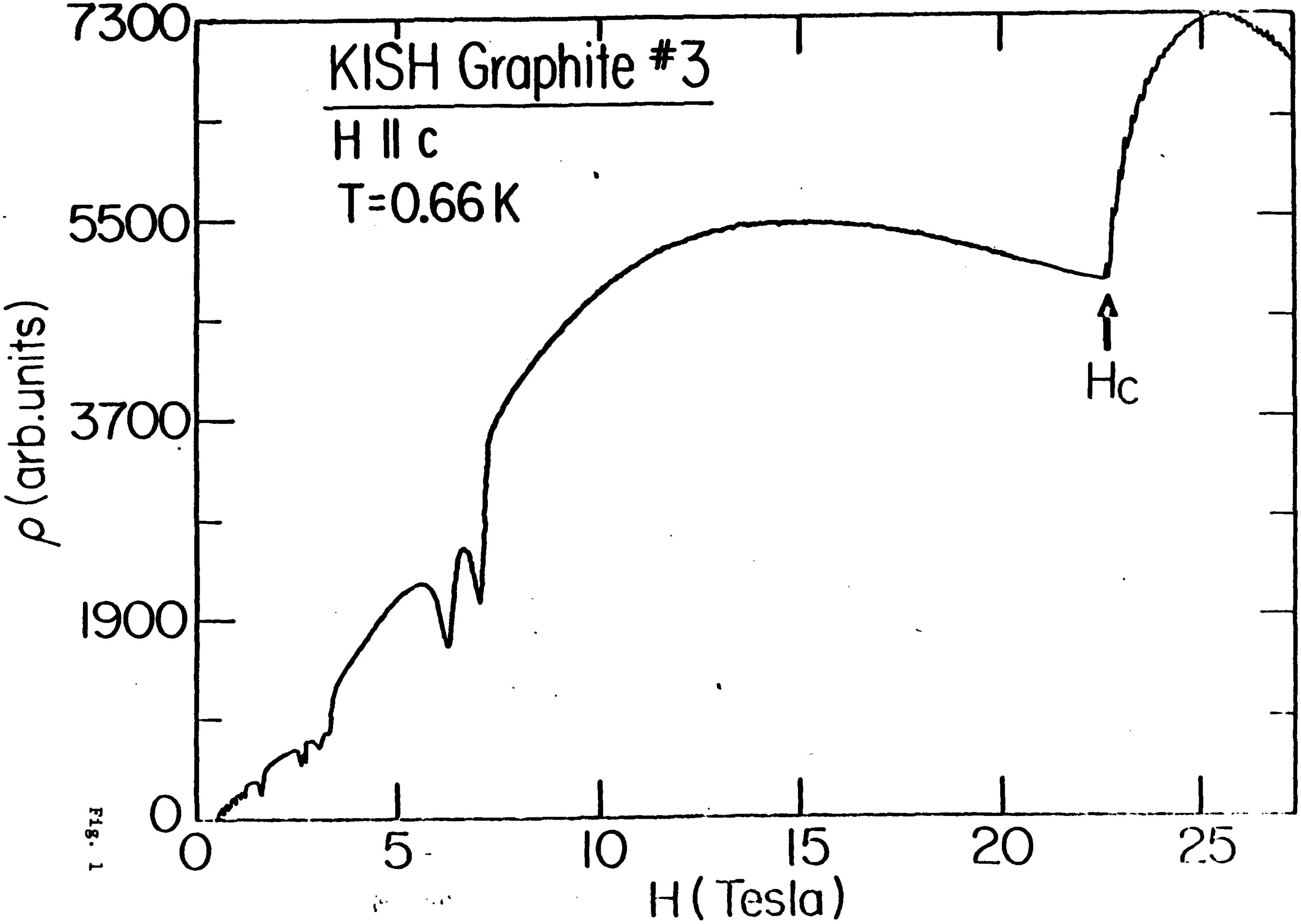
Figure captions.

Fig. 1. High field transverse magnetoresistance of kish graphite.

Fig. 2. A plot summarizing the angular dependence of the anomaly. The inset shows the temperature dependence of the field value H_c.

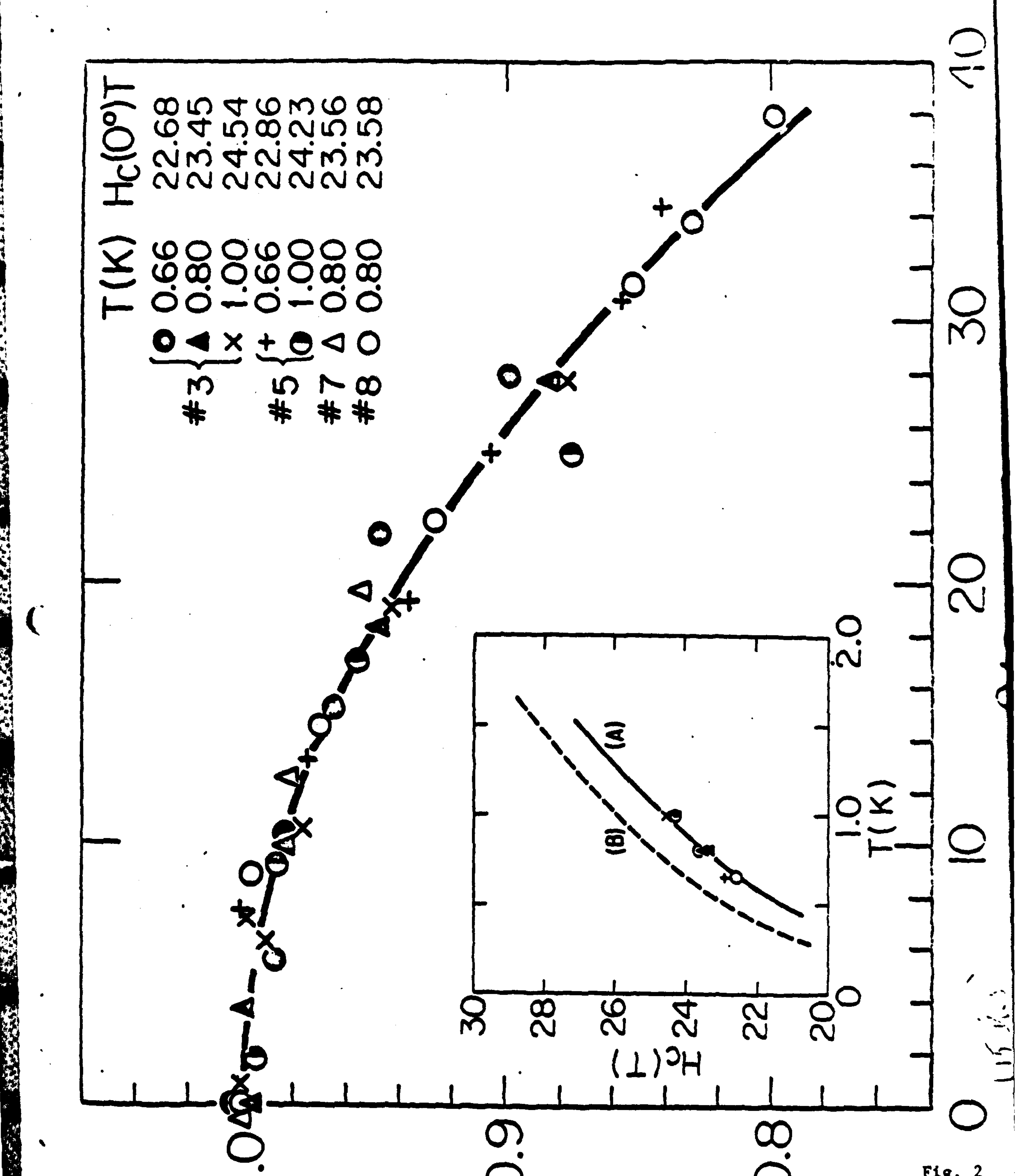
Fig. 3. Angular dependence of the magnetoresistance of graphite at constant current level and constant temperature.

Fig. 4. (a) Expanded view of the high field magnetoresistance anomaly.
(b) The corresponding oscillatory component and envelope function (see text).



KISH Graphite #3
H ll c
T=0.66 K
Hc
7300
5500
3700
1900
0
ρ (arb.units)
0
5
10
15
20
25
H (Tesla)

Fig. 1



$H_c(0°)/H_c(\theta)$
1.0
0.9
0.8
0
10
20
30
40
T(K) $H_c(0°)$T
#3 ● 0.66 22.68
▲ 0.80 23.45
x 1.00 24.54
#5 + 0.66 22.86
◐ 1.00 24.23
#7 Δ 0.80 23.56
#8 O 0.80 23.58
$H_c(T)$
30
28
26
24
22
20
0
1.0
2.0
T(K)
(A)
(B)

Fig. 2

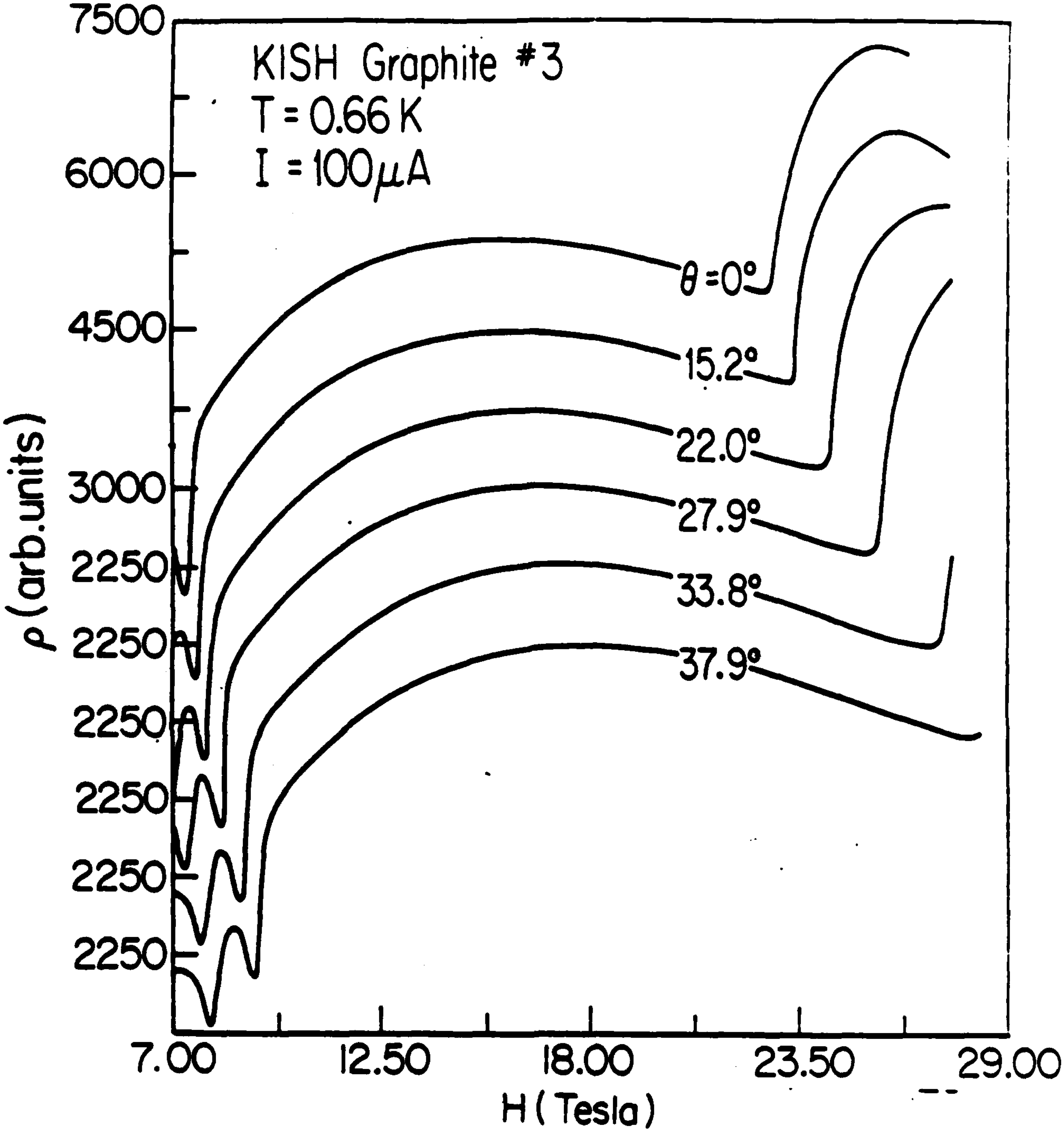
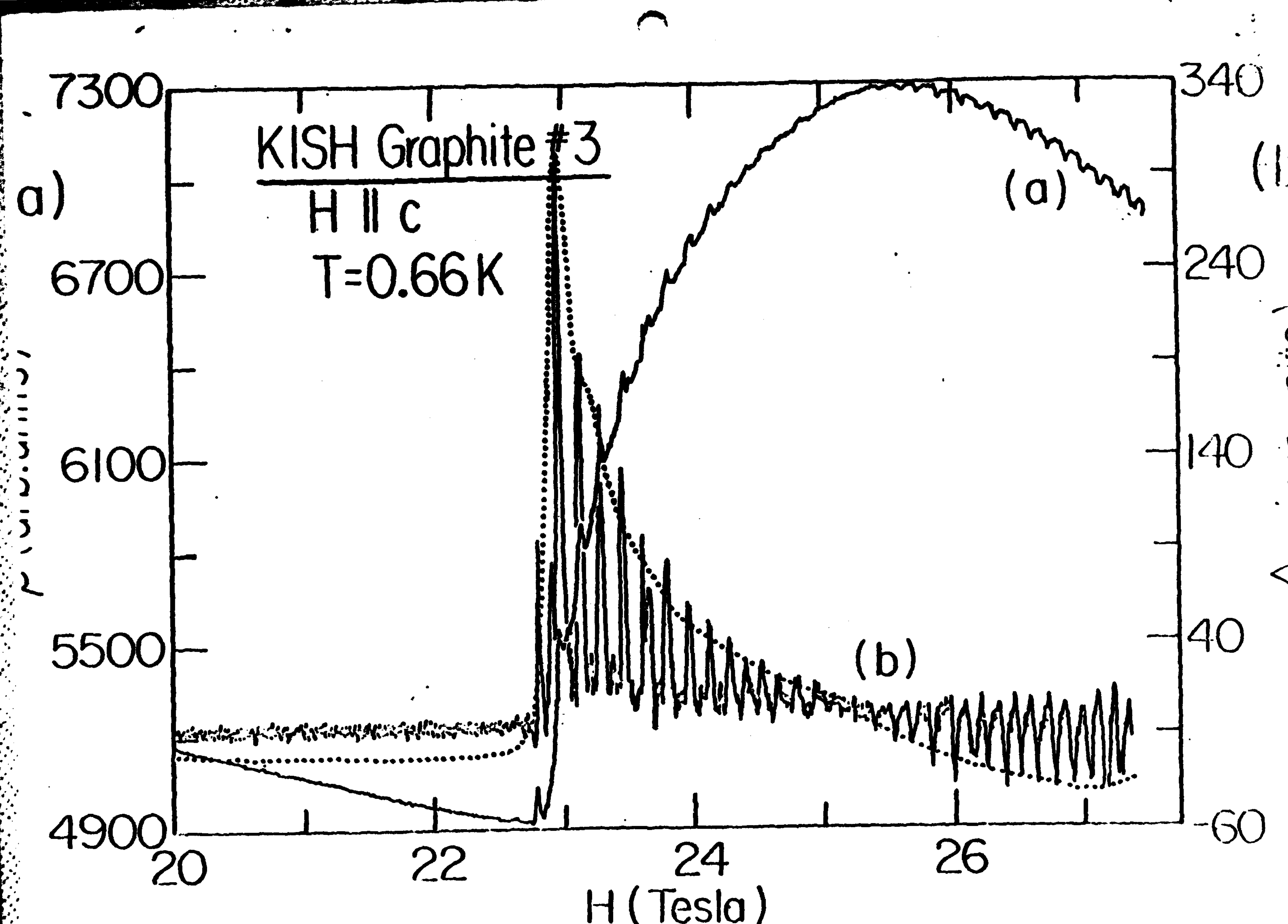


Fig. 3

$\left(\frac{7}{3.125}\right)^{-1}$ 7.25

(20 lines



a)
KISH Graphite #3
H ll c
T=0.66 K
(a)
(b)
7300
6700
6100
5500
4900
340
240
140
40
-60
20
22
24
26
H (Tesla)

Fig. 4

END

FILMED

11-83

DTIC